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Metallic Fuels: The EBR-II legacy and recent advances

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Abstract

Experimental Breeder Reactor – II (EBR-II) metallic fuel was qualified for high burnup to approximately 10 atomic per cent. Subsequently, the electrometallurgical treatment of this fuel was demonstrated. Advanced metallic fuels are now investigated for increased performance, including ultra-high burnup and actinide burning. Advances include additives to mitigate the fuel/cladding chemical interaction and uranium alloys that combine Mo, Ti and Zr to improve alloy performance. The impacts of the advances—on fabrication, waste streams, electrorefining, etc.—are found to be minimal and beneficial. Owing to extensive research literature and computational methods, only a modest effort is required to complete their development.

Keywords: metallic fuels; alloy stabilization; high burnup; electrorefining

1. Introduction

Over a span of thirty years, fuels were developed for EBR-II leading to the qualification of U-10Zr alloy (10 atom percent burnup). During this time a great deal of experience was gained in the technology and performance of metallic nuclear fuels. In addition, the operation of EBR-II was found to be safe and reliable with a compact footprint. During the 1960's, a pyrometallurgical, melt-refining process was applied to fuel alloys in their early development in the adjacent Fuel Cycle Facility (FCF) [1, 2].

During the 1990's, the FCF was renamed as the Fuel Conditioning Facility, after refurbishing it and installing equipment for a Spent Fuel Treatment Demonstration conducted by Argonne National Laboratory for the U.S. Department of Energy. In order to treat the sodium-bearing fuel, great strides were made in the science and

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technology. The processes involved were once again pyrometallurgical: molten salt electrorefining and vacuum distillation [3]. This successful demonstration required design and fabrication of engineering-scale equipment, development of criticality safety strategy [4], development of process control and monitoring [5], development of materials control methods, etc. [6, 7].

The success of the U-10Zr and other alloys, coupled with the success of the treatment demonstration, prompts the investigation and use of similar metallic fuels for burning transuranics (TRUs) to reduce the long-term environmental impact and repository loading. The potential value of improved fuel performance is recognized, with respect to safety and extended burnup, and with respect to TRU burning. For these reasons, advanced metallic fuel concepts are being researched.

2. Background

An advanced metallic fuel concept is being investigated in a collaborative effort between Idaho National Laboratory and Argonne National Laboratory. The concept incorporates major innovations that include decreased fuel smear density, annular compared to solid fuel geometries, coatings or liners on inner cladding wall, vented fuel pin, advanced fabrication methods, targeted fuel alloy additions, U-Mo based fuel alloy, and compatibility with an electrochemical fuel recycling process [8]. An INL research effort includes a focus on the latter two innovations—targeted fuel alloy additions and U-Mo based fuel alloys—to enhance the chemical stability of metallic nuclear fuels [9]. Significant improvements have the potential to extend fuel reliability to ultra-high burnup, as well as facilitate transuranic burning.

Improved chemical stability can take two forms. In one case, lanthanide fission products are known to interact strongly with cladding because of the low-melting eutectics with iron that produces fuel cladding chemical interaction (FCCI) [10, 11]. In the second case, the fuel alloy itself can interact with the cladding, especially on introducing TRUs for burning, again because of low-melting eutectics with iron [10]. This paper summarizes recent beneficial developments in fuel alloys to overcome these two types of problems, and it describes the compatibility of those alloy developments with the existing processes.

In the first case, lanthanide fission products are known to migrate to the fuel slug periphery where they can undergo FCCI after the fuel swells to close the fuel-cladding gap. A variety of evidence and analysis indicated that additives could be used to stabilize the lanthanides as intermetallic compounds, possibly to mitigate their transport, and certainly to mitigate FCCI [10, 12]. The impact of candidate additives, such as palladium and indium, on the fuel alloy is here described within the overall context of a fuel cycle.

In the second case, the addition of TRUs to the fuel alloy for burning is known to reduce the melting temperature (solidus) of the alloy, which lowers the safety margin and which can promote FCCI with alloy components [9, 13]. The U-10Zr alloy has been favored because of the steady increase in solidus temperature with higher zirconium content. The higher solidus allows plutonium and neptunium to be incorporated in the alloy, as these elements always lower the solidus and have low-melting, binary eutectics with iron. However, U-10Zr has the drawback of constituent redistribution because it does not exhibit single-phase cubic symmetry at reactor temperatures [14]. This behavior promotes anisotropic fuel swelling, and renders modeling of fuel properties difficult. On the other hand, U-10Mo alloy is single phase cubic above 550 C, has good thermal conductivity, but suffers a more severe drop in solidus on introduction of TRUs to the alloy for burning. For this reason, a superior U-10M alloy was sought, where M is some optimal combination of molybdenum, titanium, and zirconium.

While the electrorefining process has been previously described [6, 15], a brief summary is given here to help provide context for assessing the impact of the new alloy developments. Chopped segments of spent fuel are loaded into anode baskets that are then immersed into molten LiCl-KCl (eutectic ratio) at 500 C. A steel mandrel is used as the cathode during electrorefining to collect the purified uranium metal dendrites. At the anode,

therefore, uranium in the spent fuel is oxidized into the molten salt as U^{+3} while at the cathode U^{+3} is reduced to uranium metal. Active metal fission products (such as lanthanides) accumulate in the molten LiCl-KCl as their respective chlorides (e.g., $LaCl_3$). Metal fission products more noble (less electropositive) than uranium (such as Tc, Ru) are either retained in the cladding hull segments or released into the electrorefiner where they adhere to vessel components or dissolve in liquid metals. Alloy components such as zirconium can be partially oxidized to deposit with the purified uranium, or can suffer the same fate as the more noble metal fission products (such as Tc, Ru).

As mentioned, this paper examines the impacts of 1) additives to bind lanthanides as intermetallic compounds and mitigate FCCI and 2) pseudo-binary alloys involving Mo-Ti-Zr to give more uniform fuel properties while offering a good safety margin toward fuel melting. It will be seen that the desired improvements can be obtained with a modest development effort.

3. Impact of fuel alloy modifications

The impact of fuel alloy modifications should be assessed with respect to a variety of concerns. These concerns include fuel fabrication, fuel properties such as thermal conductivity and phase behavior, waste streams, and fuel recycle. These impacts have been assessed for the alloy modifications under discussion, and the impacts are found to be compatible with existing processes and beneficial overall. In general, the implementation of fuel alloy modifications can be undertaken with only a modest science-based, engineering-driven, research and development effort. Only a modest effort is needed because of the existing wealth of technical advances in metal fuel behavior and processing and because of advances in computational modeling.

In the following two sections, impact summaries are given separately first for additives (such as Pd) and second for new alloy constituents (Mo and Ti). The impacts can be related to their casting and fresh fuel behavior, their possible fates in waste streams, and their effects on neutronics. The impact on electrorefining is discussed in a combined section for all the possible fuel modifications.

3.1. Impact of additives to stabilize lanthanides fission products

Fresh fuel casting studies with U, Zr, Pd, and lanthanides confirmed initial expectations; palladium binds the lanthanides as a separate intermetallic phase and these will mitigate FCCI [10]. However, fresh fuel castings of U-15Zr (wt%) with only palladium (2-4wt% Pd, *no* lanthanides) showed that palladium lowered the zirconium content of the fuel matrix to an average of approximately 9 wt% zirconium. Similarly, when fresh fuel U-10Zr castings were produced for irradiation in the Advanced Test Reactor (ATR), the zirconium content in the fuel matrix was approximately 3-4 wt%. Consequently, a pre-alloying procedure with U and Zr was needed to keep palladium from producing high-melting Pd-Zr intermetallics, which are slow to dissolve in the melt.

In a separate effort, diffusion couple studies showed that PdNd intermetallic (i.e., 1:1 atomic ratio) does not interact with iron at 700 C while Nd interacts strongly with iron at that temperature to produce transient liquefaction and iron-rich intermetallics (eutectic temperature is 685 C) [16]. This observation is important for two reasons. First, lanthanide-based FCCI will clearly be reduced with palladium as a fuel additive. Second, since lanthanides are not soluble in the U-Zr fuel matrix, the lanthanides present in *recycled* fuel, as carryover contaminants, will phase separate during casting operations and contact cladding upon fuel swelling. As a result, FCCI can be accelerated in recycled fuel (with lanthanides) as compared to fresh fuel (without lanthanides). Palladium as an additive will therefore protect recycled fuel against accelerated, premature FCCI, in addition to permit higher burnups.

The composition range planned for the use of additive palladium is on the order of 2 to 4 wt% palladium. At approximately 1.6 wt% Pd, the palladium would be sufficient to bind lanthanides at a 1:1 stoichiometric ratio for

burnups up to approximately 8 atom%. At that burnup level, spent fuel contains on the order of 2 wt% lanthanides [10]. Since EBR-II was qualified to routine burnups of approximately 10 atom%, it can be estimated that fuel failures from lanthanide-based FCCI would be eliminated for routine burnups of approximately 20 atom% by adding approximately 2 wt% palladium to the fuel alloy. Similarly, higher burnups on the order of 25-40 atom% can be envisaged for palladium contents up to 4 wt%. Furthermore, PIE (post-irradiation examination) data for fissium fuel suggests the addition of palladium is disproportionately more effective at higher concentrations, so that a concentration of 3 wt% palladium might yield the beneficial effect to be anticipated for a larger palladium addition.

The waste streams for lanthanides may be modified by the use of palladium additive. Since the lanthanides are rendered less electropositive, a fraction of the lanthanides may not oxidize into the salt as their respective chlorides (e.g., $LaCl_3$). As a consequence, the lifetime of the molten salt for electrorefining may be extended. The intermetallic Pd-Ln compounds will therefore either be retained with the cladding hulls or be released from the cladding hulls as particulate metals, similar to noble metal fission products such as ruthenium. It is conceivable that the increase in particulate metals will assist the retention or collection of noble metals in general. In any case, any lanthanide-bearing intermetallic compounds will eventually become part of a metal waste stream that would need to be qualified.

The impact of palladium on the neutronics and reactivity of the fuel is expected to be modest. The density of palladium is relatively high at 12 g/cm^3 and the density of Zr_2Pd is approximately 10 g/cm^3, so that the volumetric effect on reactivity is less than that for zirconium alone (6.5 g/cm^3). Pd-105 has the highest cross section with an abundance of approximately 22%. In a fast reactor spectrum, the cross-section is approximately 1.5 orders of magnitude greater than that for zirconium; however, it will be used in small amounts. The cross-sections for palladium and indium are comparable; since their atomic weights are comparable and the density of indium is considerably less than the density of palladium, palladium will displace less uranium and have a smaller consequence on reactivity than indium.

3.2. Impact of alloy constituents to include molybdenum, titanium, and zirconium

The chemical stability of the fuel alloy can be enhanced with increases to the melting temperature, because melting temperatures are an indication of bond strength and melting from uranium alloys always proceeds from the body-centered cubic (bcc) phase. These facts allow direct comparisons of alloy stabilities through their melting temperatures. However, as noted, a low onset temperature for the bcc phase is desired. Consequently, a pseudo-binary alloy system, U-M, is sought that provides the desired properties—increased solidus temperature *and* decreased bcc onset temperature.

Prokoshkin and Zakharova have determined a large region of the ternary Mo-Ti-Zr system exhibits a single-phase bcc onset temperature below 600 C [17], in fact more than 25% of the ternary diagram. A bcc ternary alloy M (e.g., M = 50Mo-43Ti-7Zr, in wt %) may therefore be combined with uranium up to approximately U-10M in order to achieve a uranium alloy with the desired low bcc onset temperature and higher solidus temperature. The rationale is straightforward: 1) use Zr and Ti to raise the solidus temperature, in combination with Mo to get an optimal onset temperature for bcc phase in the ternary Mo-Ti-Zr alloy, and 2) combine with U in a U-xM pseudo-binary system, i.e., where the Mo-Ti-Zr ratios are fixed, to optimize the combined properties. Classical alloy theory was used to confirm the feasibility of this approach [18], and more importantly, recent experimental work has confirmed expectations.

Since the bcc phase field is broad, other M compositions may be selected to tune the solidus in U-10M alloys when adding TRUs for burning. While these fuel alloys appear complex and laborious to develop, they can in fact be developed in a short time. The fresh fuel portion of alloy development (possibly six researchers over three to four years) can proceed with high confidence because of the extensive research literature for all of the binary fuel alloys (U-Zr, U-Mo, and U-Ti) and considerable literature for the ternary systems. This research literature

includes phase behavior, irradiation testing, compatibility testing with cladding materials, and PIE. Furthermore, a small number of pseudo-binary systems have already been pre-selected, because the extensive literature has undergone analysis, and because computational methods (such as CALPHAD) can be applied to optimize the alloy composition using the new data.

Fresh fuel testing is planned for five or six pseudo-binary U-xM systems. From these experimental results, one composition (U-aMo-bTi-cZr) may be chosen for irradiation and PIE. A second composition, as suggested from modeling results for existing and new experimental data, may also be chosen for irradiation and PIE. This pair of alloys would comprise the uranium-only test basis. Since TRU burning is a major reason for alloy development, a second pair of alloys with TRUs could be constituted for simultaneous irradiation, i.e., U-Pu-Np-Am-M alloys, where once again two compositions of M (two sets of a, b, and c values) are determined with the first composition determined by experiment and the second suggested by modeling. It may be seen, therefore, how optimized, high burnup alloys that incorporate TRUs for burning can be down-selected in approximately eight to ten years.

The negative impact of the alloy modifications is expected to be minimal, and the overall impact is beneficial. Casting should be little affected, based on the experience with the binary systems. It may be necessary to perform some pre-alloying step to produce a more homogeneous alloy composition. Thermal conductivity can be expected to be comparable or better than U-10Zr, because of the presence of molybdenum. Likewise, fuel swelling and constituent redistribution are expected to be improved compared to U-10Zr, because of the sizable increase in the fraction of cubic phase. Of major importance for TRU burning fuels, the safety margin toward fuel melting will be increased compared to a U-Mo alloy with TRUs. The fast reactor neutronics penalty for molybdenum is substantial in comparison to zirconium (but less than a factor of ten); however, the change in reactivity can be accommodated in a fast reactor with core size and fissile content (U-xMo, with x = 7-10 wt%, was used in both FERMI and Dounreay fast reactors). The cross section for titanium in fast reactors is comparable to zirconium, and the titanium cross section is less than zirconium for most of the spectrum between 0.2 and 3 MeV.

The impact on the waste streams should be minimal. The waste stream already needs to accommodate Tc-99, because U-10Zr at 8 atom percent burnup contains 0.17wt% (0.34 atom%) technetium [10]. The Tc-99 generated from Mo-98 in the fuel alloy therefore does not introduce a new waste stream. Otherwise, the titanium and molybdenum should behave similar to zirconium; electrorefining with a U-10M alloy is discussed in the next section.

3.3. Expected electrorefining behavior for new alloy components

Table 1 lists an abbreviated electromotive (emf) series for fuel components in LiCl-KCl eutectic at 450 C [19]. For this series, neodymium is the most electropositive and ruthenium is the least electropositive. Thus, proceeding down the table the relative stability of the chlorides is ranked, with $NdCl_3$ being the most stable chloride (with $NdCl_3$ stability comparable to the chlorides of the other lanthanide fission products). On account of the stability of the lanthanide chlorides, these fission products accumulate in the molten LiCl-KCl salt, which contributes to their disposition in the ceramic waste form. The relative abundance of chemical species present in irradiated EBR-II fuel has been previously given [6, 10].

The electrorefining behavior of zirconium in the fuel alloy has been described, with regard to its oxidation [20], deposition with uranium at the cathode [21, 22], and spurious deposition of zirconium on the exterior of the cladding [23]. Depending on the process conditions, it is possible to electrochemically oxidize zirconium from the steel cladding and the extent of oxidation can be controlled. Since the stability of the chlorides of titanium is comparable to the chlorides of zirconium in this system, the electrorefining characteristic of titanium is expected to be comparable to zirconium, and likewise controllable.

The emf for ruthenium is comparable to those for molybdenum and palladium. It has been shown that ruthenium can be retained with the cladding, and that it can contaminate electrodeposited uranium as a consequence of convection of fine particulates [24, 25]. However, palladium should be chemically bound with the lanthanides [10, 16], and the lanthanides should be more difficult to oxidize to their respective chlorides, by the lowering of their free energy upon formation of the intermetallic compound. This result could benefit the ceramic-salt waste streams, as the lanthanides would not accumulate in the eutectic LiCl-KCl as rapidly as compared to absence of palladium.

Molybdenum will not electrochemically oxidize in the electrorefining process, and its presence is expected to aid the retention of zirconium and titanium. However, this expected behavior needs to be demonstrated for an alloy composition to be decided, and the tests could be conducted initially using unirradiated, depleted uranium alloys. The presence of molybdenum could slow down and impede the electrochemical oxidation of the last few percentage points of uranium, as zirconium does, and this behavior will also have to be evaluated.

Table 1. Electromotive Force Series for Selected Elements in Eutectic LiCl-KCl at 450 °C [19]

Oxidation / Reduction Couple	E°M (Pt) (V)	$E°_m$ (Ag) (V)
Nd (III) / Nd (0)	-2.819	-2.097
U (III) / U (0)	-2.218	-1.496
U (IV) / U (0)	-1.950	-1.230
Zr (IV) / Zr (II)	-1.864	-1.153
Zr (IV) / Zr (0)	-1.807	-1.088
Zr (II) / Zr (0)	-1.75	-1.02
Ti (II) / Ti (0)	-1.74	-1.01
Ti (III) / Ti (0)	-1.60	-0.88
Ti (IV) / Ti (0)	-1.486	-0.767
Cr (II) / Cr (0)	-1.425	-0.698
Ti (III) / Ti (II)	-1.32	-0.61
In (I) / In (0)	-1.210	-0.467
Fe (II) / Fe (0)	-1.172	-0.445
Mo (III) / Mo (0)	-0.603	+0.119
Pd (II) / Pd (0)	-0.214	+0.513
Ru(III) / Ru (0)	-0.107	+0.615

3.4. Fuel alloys irradiated at the Advanced Test Reactor

A subset of the alloys that are being researched has recently been irradiated at the Advanced Test Reactor at the Idaho National Laboratory [26]. Fuel features being examined include smear density, annular versus rod geometries, and the use of palladium as an additive. However, only binary U-Zr and U-Mo alloys are being examined with respect to these features. Future tests are expected with the novel alloys involving uranium with ten weight percent of (aMo-bTi-cZr), where a, b, and c are the weight fractions to be selected after characterizing a few different compositions using x-ray, microstructural and thermal methods.

4. Conclusion

Some of research efforts at the Idaho National Laboratory on new uranium fuel alloys are directed toward improving the chemical stability of the fission products to mitigate FCCI and toward improving the thermodynamic stability of the fuel phases. In comparison to the base alloy of U-10Zr, possible new alloy components include palladium and indium (for improved FCCI stability), and titanium and molybdenum (for improved thermodynamics). Palladium or indium would be used for binding the lanthanide fission products as intermetallics, to mitigate FCCI. Titanium and molybdenum would be used with zirconium as alloy constituents (fuel matrix) to improve fuel properties, particularly increased melting temperature with a decreased onset temperature for the cubic phase. The increased melting temperature is particularly important for transuranic burning. Plutonium and neptunium always lower the alloy melting temperature and this lowering must be compensated for to have a reasonable safety margin.

The electromotive series for these elements in molten LiCl-KCl eutectic indicates that their presence should not unduly complicate the electrorefining process and that they should behave comparably to zirconium and iron (i.e., titanium and indium) and to ruthenium (i.e., molybdenum and palladium). Analysis of the extensive research literature, recent experimental results, and the assessed impacts of the alloy modifications give high confidence that a superior alloy can be successfully developed with modest effort in a reasonable time. These alloys can be reasonably expected to bring performance improvements, such as high burnup, and permit transuranic burning with an improved safety margin toward fuel melting.

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